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DOI:

[10.1016/j.envint.2020.106041](https://doi.org/10.1016/j.envint.2020.106041)

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Document Version

Publisher's PDF, also known as Version of record

Citation for published version (Harvard):

Sharkey, M, Harrad, S, Abou-Elwafa Abdallah, M, Drage, DS & Berresheim, H 2020, 'Phasing-out of legacy brominated flame retardants: the UNEP Stockholm Convention and other legislative action worldwide', *Environment International*, vol. 144, 106041. <https://doi.org/10.1016/j.envint.2020.106041>

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Review article

Phasing-out of legacy brominated flame retardants: The UNEP Stockholm Convention and other legislative action worldwide

Martin Sharkey^{a,*}, Stuart Harrad^b, Mohamed Abou-Elwafa Abdallah^{b,c}, Daniel S. Drage^b, Harald Berresheim^a

^a School of Physics, National University of Ireland Galway, University Road, Galway H91 CF50, Ireland

^b School of Geography, Earth, and Environmental Sciences, University of Birmingham, Birmingham B15 2TT, United Kingdom

^c Department of Analytical Chemistry, Faculty of Pharmacy, Assiut University, 72516 Assiut, Egypt



ARTICLE INFO

Handling Editor: Heather Stapleton

Keywords:

Persistent organic pollutants
Brominated flame retardants
Concentration limits
Legislation
Circular economy
Flame retardancy standards

ABSTRACT

Due to their toxicity and persistence, several families of brominated flame retardants (BFRs) have been listed as persistent organic pollutants (POPs) in the Stockholm Convention, a multilateral treaty overseen by the United Nations Environment Programme. This treaty mandates that parties who have signed must take administrative and legislative actions to prevent the environmental impacts that POPs pose, both within their jurisdictions and in the global environment. The specific BFRs listed in the Stockholm Convention are Polybrominated Diphenyl Ethers (PBDEs), Hexabromocyclododecane (HBCDD), and Hexabromobiphenyl (HBB), chemicals which must therefore be heavily restricted within the jurisdictions of the signatories. As an example, within the EU, hexabromobiphenyl (HBB), the PBDE commercial mixtures, and HBCDD are almost entirely prohibited in terms of both production and use in commercial goods. Waste articles containing excess concentrations of these BFRs are similarly restricted and must be disposed of in a manner that destroys or irreversibly transforms the BFR in question. In some cases, specific exemptions for these limits are defined by the Convention for certain parties: for example, Penta- and Octa-BDE can be present in waste materials for recycling until 2030, while Deca-BDE can be applied to some aviation and automotive applications until 2036. However, in such cases, very specific criteria and guidelines apply for their use and/or production. Worldwide, China, Japan, India, and the United States of America have made significant advances in the regulation of POPs, in line with the provisions of the Stockholm Convention. China has established concentration limits for Penta- and Octa-BDEs in electronic goods. It is also currently availing of an exemption to allow for the use of HBCDD and has not yet ratified the Convention with regards to Deca-BDE. Japan meanwhile has classified HBB and Penta-/Octa-BDE compounds as Class I Specified Chemical Substances which virtually prohibits the manufacture, import, and use of these chemicals in all applications. India has banned the manufacture, trade, import, and use of HBB, HBCDD and some PBDEs, and has established concentration limits for all PBDEs in certain electrical goods. Finally, the United States has no federal mandate for the restriction of POPs and has not ratified the annexes to the Convention requiring them to do so. However, thirteen states have implemented their own state-wide concentration limits on a variety of flame retarding chemicals in various commercial applications. Though these limits worldwide are a very positive step for the removal of POP-BFRs from the environment, the increased use of replacement flame retardants renders such legislation only partially effective. The lack of effective screening mechanisms in waste management facilities means that BFR-treated plastics can be inadvertently recycled and remain in circulation. The rise in the use of novel BFRs (NBFRs) can furthermore hinder screening methods currently being developed and the additives themselves may pose similar issues to their predecessors owing to their similar chemical properties. Thus, restrictions on current BFRs will result in the use of new flame retardants, which may in turn be banned and replaced once again. Further research into and development of methods to screen for hazardous chemicals in end of life materials is therefore of the utmost importance. This must be coupled with pro-active legislation that eliminates the need for using such persistent and potentially harmful chemicals in the future.

Abbreviations: MEA, Multilateral Environmental Agreement; REIO, Regional Economic Integration Organisation; UTC, Unintentional Trace Contaminant; UNEP, United Nations Environment Programme.

* Corresponding author.

E-mail address: martin.sharkey@nuigalway.ie (M. Sharkey).

<https://doi.org/10.1016/j.envint.2020.106041>

Received 21 June 2020; Received in revised form 31 July 2020; Accepted 4 August 2020

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1. Introduction

1.1. The United Nations Environment Programme (UNEP)

The United Nations Environment Programme is “... *the leading global authority that set out the global environmental agenda*” (UNEP, 2008d). Since its inception, UNEP has instigated a number of multilateral environmental agreements (MEAs) which are “*legally binding international instruments through which national Governments commit to achieving specific international goals*”. Some examples of these MEAs are: the Basel Convention on the control of transboundary waste and its disposal; the Rotterdam Convention on the prior informed consent procedure for certain hazardous chemicals and pesticides; and the Stockholm Convention on persistent organic pollutants (POPs). These MEAs, though stated to be legally binding, only apply to those countries and Regional Economic Integration Organisations (REIOs – e.g. the European Union) who have agreed to be bound by said agreement. The inherent benefits for compliance with agreements can be seen as largely moral or political, i.e. being for the cause of environmental protection for future generations or improving the health and lifestyle for the population of a given country/REIO. More tangible incentives for adopting an MEA have also been introduced and are generally specific to each agreement. For example, certain MEAs prevent or have specific guidelines for trade with non-MEA states, thus hindering international trade. Funding can also be available to states for achieving goals in-compliance with an MEA. Multilateral networks such as these furthermore foster closer ties between co-signatories which improves international cooperation and fosters the creation of bilateral or multilateral agreements for achieving shared goals or for trading technology and resources (UNEP, 2006a). Measures have furthermore been taken by UNEP to “strengthen and upgrade” their ability to require compliance with MEAs. A resolution by the UN General Assembly introduced an initiative to “*decouple economic growth from unsustainable use of natural resources*”, an action which aims to improve the rate of compliance with environmental agreements among UN member-states by reducing the economic burden associated with environmental conservation (UNEP, 2006a, UNEP, 2013b).

1.2. The Stockholm Convention on persistent organic pollutants

The Stockholm Convention is an international treaty which primarily aims to “... *protect human health and the environment from persistent organic pollutants*” (UNEP, 2008c). The Convention was instigated by the governing council of UNEP in 1995 and entered into force in 2004 after the fiftieth “instrument of ratification” was received, thus meeting the quorum of UN member-states being party to the Convention. The Convention is predicated on Principle 15 of the Rio Declaration on Environment and Development (UN, 1992) which states: “*In order to protect the environment, the precautionary approach shall be widely applied by states according to their capabilities. Where there are threats of serious or irreversible damage, lack of full scientific certainty shall not be used as a reason for postponing cost-effective measures to prevent environmental degradation.*”

According to UNEP, these POPs are chemicals which have “... *a particular combination of physical and chemical properties such that, once released into the environment, they (i) remain intact for exceptionally long periods of time, (ii) become widely distributed throughout the environment as a result of natural processes involving soil, water, and, most notably, air, (iii) accumulate in the fatty tissue of living organisms including humans and are found in higher concentrations at higher levels in the food chain, and (iv) are toxic to both humans and wildlife*” (UNEP, 2008c). In general, a chemical is designated as a POP when it has an environmental half-life greater than 2 months, a bio-accumulation factor greater than 5,000, has an atmospheric half-life exceeding 2 days, and there is documented evidence of human or environmental toxicity (see Fig. 1 for further details and other criteria). Such chemicals recognised as POPs become the purview of the Stockholm Convention, whose mandate is to study,

monitor, and ultimately restrict the use and prevalence of relevant chemicals in the environment.

An initial list of 12 POPs were included in the Convention, with scope for the inclusion of more hazardous chemicals should its Conference of Parties (CoP) deem such inclusions a necessity (UNEP, 2008a). Where research has shown that a chemical may fulfil the criteria for being considered a POP, the Convention's POPs Review Committee is tasked with assessing the hazards posed by the chemical in question in order to make recommendations to the CoP regarding its suitability for inclusion. If subsequently listed, states who are signatories of the treaty (i.e. a “party” to the Convention) are required to take legal and administrative action to (i) reduce or eliminate releases from intentional production and use, (ii) restrict the import/export of POPs and articles treated with POPs as required, and (iii) notify the Secretariat of the Convention any “specific exemptions” to the Convention which are required by the party (UNEP, 2009).

These exemptions are criteria for the production and/or use of POPs listed in the Convention, which parties may qualify for if sufficient reasoning is provided. These exemptions (if any have been specified) are unique to each POP but all have an expiration date of not more than five years after entry into force of the Convention for that specific POP, unless otherwise applied for and granted to a party (UNEP, 2009). However, those listings only include countries which have ratified the Convention and officially stated that they are unable to comply with the restrictions. Though there are currently 152 signatories to the Convention, several parties have yet to ratify the tenets therein, i.e. their intention to abide by the restrictions posed by the Convention. At the time of writing, those signatories which currently have no national restrictions in place for the production or use of any POP are Brunei, Haiti, Israel, Malaysia, and the United States of America (UNEP, 2020a).

2. Legacy BFRs and the UNEP Stockholm Convention

2.1. Brominated flame retardants in the Stockholm Convention

The widespread use of plastic-based consumer articles since the mid-twentieth century was accompanied by a rise in fire-safety concerns due to said polymers being inherently flammable (Junod, 1976). This resulted in the creation of a new class of chemicals known as flame retardants (FRs), the primary function of which was to prevent combustion of a material or hinder a fire's propagation and progression. Among the most common of these FRs used in household polymers were phosphorous- and halogen-based FRs: the former, reportedly being more typically applied to textile coating agents; the latter, being widely used in the hard plastics of consumer articles (van Esch, 1997, La Guardia and Hale, 2015).

Over the last few decades, the brominated variants of these halogenated FRs have been the subject of intense scrutiny due to their toxicity to humans, their extensive application in consumer articles, and their increasing prevalence in the environment (Eljarrat and Barceló, 2011). Built upon the wealth of research on these chemicals, UNEP have adopted measures to reduce the prevalence and ubiquity of brominated flame retardants (BFRs) currently in circulation and in the environment at large. Specifically, these regulated BFR families are polybrominated biphenyls (PBBs), polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD), all of which are applied to consumer articles in the form of “commercial mixtures” (La Guardia et al., 2006) which are dominated by certain congeners or isomers of said BFRs. For example, a common commercial mixture from the PBDE family is “Penta-BDE”, which is dominated by tetra- and pentabromodiphenyl ether congeners and was used in polyurethane foams in furniture and vehicles (La Guardia et al., 2006, European Commission, 2011).

To date, there are five specific groups of BFRs listed in the Convention, all of which are listed in Annex A (Elimination). These are:

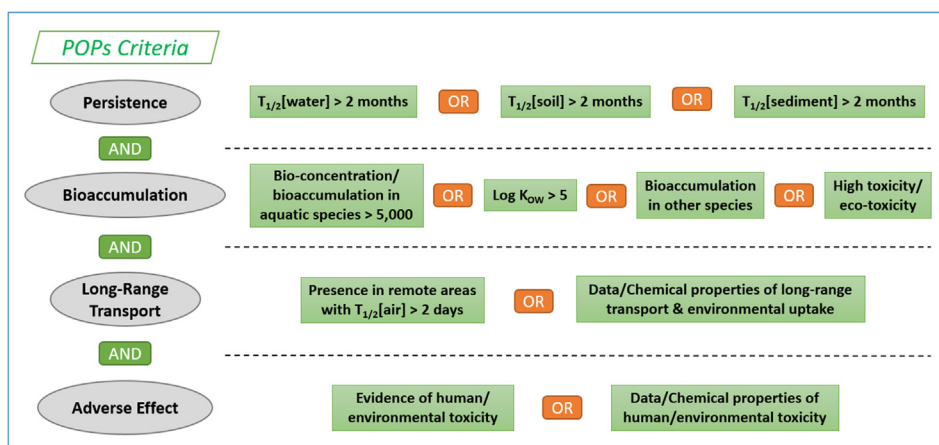


Fig. 1. Overview of POPs criteria as outlined in Annex D of the Stockholm Convention (UNEP, 2009) (Figure adapted from Sharkey (2019)). $T_{1/2}[X]$ = environmental half-life of a chemical in a given medium; $\text{Log } K_{OW}$ = Octanol/Water partitioning coefficient.

hexabromobiphenyl (HBB); HBCDD; commercial Octa-BDE; commercial Penta-BDE; and commercial Deca-BDE (UNEP, 2009, UNEP, 2013a, UNEP, 2015, UNEP, 2017a). The Stockholm Convention refers to specific chemical substances related to these BFRs based on their Chemical Abstracts Service (CAS) registry number in addition to some non-specific references, namely "... other hexa- and heptabromodiphenyl ethers present in commercial Octa-BDE" and "... other tetra- and pentabromodiphenyl ethers present in commercial Penta-BDE" (UNEP, 2009) (see [Supplementary Information – S2](#) for a full overview).

2.2. Restrictions and exemptions for PBDEs

Penta- and Octa- BDE commercial mixtures were listed in the Stockholm Convention in 2004, while Deca-BDE – used as a replacement for Penta-/Octa-BDEs in several applications – was listed much later, in 2019. For those congeners within the Penta-BDE and Octa-BDE commercial mixtures, there are no specific exemptions allowed for the production of these substances. There is, however, an exemption pursuant to the use of these chemicals (only for parties who register for it), specifically with regards to the recycling of contaminated waste articles. This exemption allows a registered party to allow the recycling of articles that contain or may contain these substances, as well as the use and/or final disposal of new articles containing the recycled materials. They must furthermore ensure that these new articles are disposed of in an environmentally sound manner, as well as take steps to prevent articles (both pre- and post-recycling) being exported which contain banned substances at concentrations exceeding those permitted for the sale, use, import, or manufacture within their jurisdictions (UNEP, 2020c, UNEP, 2020d).

States which, as of the time of writing of this article, avail of this exemption for the use of Penta-BDE are Brazil, Japan, Turkey, and the Republic of Korea. These exemptions were largely sought as BFR-treated materials are currently in circulation within their industries and it is not economically feasible to prevent PBDE-treated materials entering the recycling system. States which avail of the exemption for Octa-BDE include the four exempted for Penta-BDE, as well as Cambodia, each for similar reasons as for Penta-BDE (i.e. contamination of waste in recycling processes). Ordinarily, the allowance for this exemption would expire five years after the date of entry into force of the restriction on the chemical, i.e. by 2009 in these cases. However, the Convention's CoP can extend the expiry date for specific restrictions beyond the initial five year period. These dates have been extended for all of the parties currently listed in the Register of Exemptions for both Penta-BDE and Octa-BDE (where applicable). There are no current revised timelines for these parties as to when these exemptions will expire, thus the only expiry date officially in place will be in 2030 when

said exemptions will no longer be available (UNEP, 2020c, UNEP, 2020d).

Deca-BDE is the most recent BFR to be listed in Annex A of the Stockholm Convention and its accompanying CoP decision (UNEP, 2017a) outlines the specific exemptions allowed for parties which register and qualify for them. These exemptions are, broadly speaking, for: specific parts for certain legacy vehicles which have ceased mass-production, as well as spare parts for said vehicles; materials for the building of specified types of aircrafts and their spare parts; additives for plastic housings and parts for electrical and electronic equipment (EEE) required to comply with fire retardancy standards at concentrations lower than 10% by weight; textiles (excluding clothing and toys) requiring anti-flammability characteristics; and polyurethane foams for building insulation. The parties who have applied for these specific exemption(s) include Brazil (import for use in vehicles), the EU (for production and use in vehicles, aircrafts, and additives in plastics for heating applications), Iran (uses in textiles and coatings), New Zealand (import for use in vehicles and aviation spare parts), the Republic of Korea (production for additives in plastic housing and use in vehicular and aviation spare parts), and Switzerland (production and use for vehicle and aviation spare parts) (UNEP, 2019). These exemptions can be withdrawn at any time by the parties themselves, but will regardless expire by 2036 (UNEP, 2017a).

2.3. Restrictions and exemptions for HBCDD

The restriction of HBCDD came into effect on the 26th of November 2014, which included a specific exemption related to both its production and its use. This exemption states that HBCDD can only be produced and used in expanded and extruded polystyrene foams for use in buildings, provided that said foams are clearly labelled and/or identifiable as containing HBCDD throughout its lifetime. At the time of writing, China is the only party to avail of the exemption for the production of HBCDD, while the Republic of Korea as well as China both avail of the exemption for its use. Both parties are subject to the five-year expiry date of the exemption, these being the 27th of October 2020 for the Republic of Korea and the 26th of December 2021 for China (UNEP, 2020b).

2.4. Restrictions and exemptions for HBB

HBB is a legacy BFR used extensively alongside PBDEs and HBCDD (see [Section 2.1](#)). HBB was listed in the Convention in 2004 along with Penta- and Octa-BDE. Unlike the other BFRs listed in the same year, there were no specific exemptions accompanying the listing of this chemical, either in its production or use, for any party.

3. Legacy BFRs and the EU

3.1. EU regulations governing legacy BFRs

Within the EU, the primary regulation relating to the governance of POPs is Regulation 2019/1021 on persistent organic pollutants (EU, 2019). This regulation stems directly from the Stockholm Convention as well as the MEA on Long-Range Transboundary Air Pollution of Persistent Organic Pollutants, from which it identifies POPs in need of restriction and/or prohibition within the EU. The second major ruling which governs the use of POPs in the EU is Directive 2011/65/EU on the “restriction of the use of certain hazardous substances in electrical and electronic equipment” or “RoHS” for short (EU, 2011). The POPs regulation and RoHS directive operate together and define concentration limits for various hazardous substances (including BFRs) in consumer articles. The first set of limits these regulations define are “unintentional trace contaminant” (UTC) levels for BFRs in consumer articles entering the market, whether newly manufactured or made from recycle. The second set of limits, govern concentrations in articles entering the waste stream, concentrations above which said waste cannot be disposed of by conventional means (e.g. landfilling or recycling) and must be disposed of via specialised methods. Three such disposal methods are defined in the regulation: physicochemical treatment, incineration on land, and use as a fuel or other means to generate energy. In addition to these disposal methods, pre-treatment of the waste which isolates the hazardous substance from the article(s) is permitted which in turn allows said articles to be disposed of in the conventional means. However, this is only permitted provided that the isolated substance is disposed of via one or more of the three specialised disposal methods outlined above (EU, 2019).

POPs and RoHS work in tandem with Regulation 1907/2006 concerning the “Registration, Evaluation, Authorisation and Restriction of Chemicals” (REACH) (European Commission, 2006). Due to the Stockholm Convention stipulating that a listed POP may be used in specific cases for a period of time, such specific exemptions must be submitted to and authorised by REACH. The sections below outline the purview and effect of these legislations as pertains to the each listed BFR (Table 1).

3.2. Current restrictions on polybrominated diphenyl ethers

Each of tetra-BDE, penta-BDE, hexa-BDE, hepta-BDE, and deca-BDE are listed individually in the POPs regulation (EU, 2019) with UTC concentration limits established for these substances placed on the market. There is then a separate listing for the sum of the listed PBDEs in articles and mixtures placed on the market, as well as for the concentrations allowable in waste (see *Supplementary Information – S3* for further information). For each of the individual PBDEs listed, the UTC limit in substances is 10 mg/kg (0.001%) meaning that the presence of any one of these homologue groups above this limit in a substance cannot be placed on the market. The UTC limit in place for the sum of PBDEs in articles and mixtures is currently 500 mg/kg (with the exception of Deca-BDE for which some exemptions exist – subject to review by 16th July 2021). Deca-BDE is currently the only PBDE which has specific exemptions allowed for its production and use; as such, it is also listed in REACH, outlining the specific exemptions for its production/use and defining a market UTC limit of 1,000 mg/kg (0.1%) in substances, mixtures and articles (European Commission, 2006). Though the UTC limits defined by the POPs and REACH regulations differ, they operate in tandem with one another; therefore, the lower limit of 0.05% is the current limit on deca-BDE permissible in mixtures or articles placed on the market and 0.001% for deca-BDE in substances. These limits defined above for PBDEs by the POPs and REACH Regulations do not extend indiscriminately across all consumer articles. Both specifically state that these particular limits do not apply to EEE within the scope of the RoHS Directive. This concentration limit for EEE

Table 1
Overview of BFRs listed in the Stockholm Convention, listing: the year each chemical family was introduced; whether specific exemptions for use and/or productions exist and which countries/REIOs avail of them; and the year those exemptions will expire. Abbreviations “u” and “p” refer to use and production exemptions respectively; each country/REIO listed is availing of an exemption for the use and/or production of the relevant BFR.

BFR	Year of Listing	Exemption(s) for Use?	Exemption(s) for Production?	Country (u/p)*	Exemption Expiry Year
tetra/penta-BDEs	2004	Y	N	Brazil (u), Japan (u), Turkey (u), Republic of Korea (u)	2030
Hexa/hepta-BDEs	2004	Y	N	Brazil (u), Japan (u), Turkey (u), Republic of Korea (u), Cambodia (u)	2030
Deca-BDE	2019	Y	Y	Brazil (u), EU (u/p), Iran (u), New Zealand (u), Republic of Korea (u/p), Switzerland (u/p)	2036
HBCDD	2014	Y	Y	China (u/p), Republic of Korea (u)	2021
HBB	2004	N	N	none	2009

is set at 0.1% by weight for the sum of all PBDE congeners in a given article or its components that is placed on the market (EU, 2011).

With the recast of the POPs regulation came a limit for the concentrations of PBDEs in all waste articles (including EEE). This limit is also 1,000 mg/kg (0.1%) for the sum of all PBDE congeners in a given article, beyond which the waste item is considered “hazardous” and cannot be disposed of by conventional methods (EU, 2019). Additionally, a concentration limit of 10,000 mg/kg is specified for the sum of the aforementioned PBDE congeners in waste from thermal processing/incineration, metallurgical processing, construction and demolition, and industrial processes (EU, 2019). The previous POPs Regulation (no longer in effect since the 2019 recast) defined several concentration limits for articles placed on the market made from recycled materials. These limits were 1,000 mg/kg (0.1%) for each of the tetra-, penta-, hexa-, or hepta-BDE homologues (European Commission, 2004, EU, 2010); however, the revised regulation (EU, 2019) no longer has any such distinction, meaning the 10 mg/kg UTC now applies to all substances placed on the market and the 500 mg/kg UTC limits for the sum of PBDEs in mixtures and articles placed on the market, whether it is made from recycled materials or not (with the exception of EEE within the scope of the RoHS Directive).

3.3. Current restrictions on hexabromocyclododecane

HBCDD has established concentration limits for both articles placed on the market as well as for the articles entering the waste stream. The limit for disposal by conventional means is defined at 1,000 mg/kg (0.1%). This concentration limit is, unlike for PBDEs and HBB, also maintained at the same level for wastes from thermal processing/incineration, metallurgical processing, construction and demolition materials, and industrial applications (EU, 2019). The UTC concentration for HBCDD is meanwhile set at 100 mg/kg, above which an article cannot be placed on the market. Up until late 2019, the production and use of HBCDD was allowable in certain applications (namely for use in household insulation). However, said exemption expired as of the 26th of November 2019, and its use is no longer permitted within the EU.

3.4. Current restrictions on hexabromobiphenyl

HBB was the first POP-BFR to be listed in the original EU POPs regulation and its use in commercial applications has largely been phased-out. As such, no UTC was defined for HBB in articles entering the market and comparatively low concentration limits of 50 mg/kg was defined for consumer articles entering the waste stream, and 5,000 mg/kg for wastes from thermal processing/incineration, metallurgical processing, construction and demolition, and industrial processes (EU, 2019). Finally, a special case to note regarding HBB is on the use of all polybrominated biphenyls (PBBs) in electrical and electronic equipment (as defined by the RoHS directive) placed on the market, which is allowable to a concentration of 0.1% by weight (EU, 2011) (an overview of all POP-BFR restrictions within the EU is summarised in Table 2).

4. Legacy-BFRs in other countries/REIOs

4.1. Major global electronic and textile waste producers

Electronics and textiles are the major commercial sectors in which large quantities of BFRs have reportedly been used, with construction & demolition and vehicular waste also being important (Eljarrat and Barceló, 2011). The seven largest producers of e-waste worldwide are the EU (10.07 Mt), China (7.21 Mt), USA (6.30 Mt), Japan (2.14 Mt), India (1.96Mt), Brazil (1.53), and Indonesia (1.27 Mt) (Fig. 2) (Baldé et al., 2017). With regards to textiles, China, the EU, USA, and India are among the greatest manufacturers/exporters of textiles worldwide, as well as the largest producers of textile waste annually (Statistica, 2018,

Global Recycling, 2019, FashionUnited, 2020). Each of these states are signatories to the Stockholm Convention (though USA is not an official party to the convention). However, the EU is currently the only state to have enacted provisions on all POP-BFRs with regards to waste and recycling of materials (Table 2). While most of the other major global producers have ratified the Convention's provisions to some degree, not all have adopted the provisions with regards to all POP-BFRs, and few have established *de jure* concentration limits on the levels of POP-BFRs permissible in marketable or waste plastics (Table 3).

4.2. North and South America

Brazil is currently availing of several exemptions for the use of PBDEs in certain applications (see Section 1.2), exemptions which will expire in 2030 for Penta-/Octa-BDEs and 2036 for Deca-BDE. However, there is currently no specific legislation within Brazil limiting the concentrations of PBDEs or HBCDD in commercial or waste goods, neither is there a prohibition on the import or export of these BFRs (Rodrigues et al., 2015). Both Brazil and the United States have disclosed that there is no production or use of hexabromobiphenyl in their respective countries (UNEP, 2006b). However, in contrast to Brazil, the United States has yet to adopt any of the provisions of the Stockholm Convention regarding BFRs (USDOS, 2019).

While the United States has no restrictions on BFRs at the federal (nationwide) level, thirteen states have implemented restrictions which include some limitation on the use/presence of BFRs in certain commercial goods (Table 4). Currently, Hawaii, Illinois, Michigan, and New York only include Penta- and/or Octa-BDE in their restrictions, but applied to all commercial goods entering the market (with the exception of shipping pallets in some cases). Some states have gone further still in recent years, outlawing the use of Deca-BDE, HBCDD, or all halogenated flame retardants in certain commercial goods such as children's products and upholstered furniture. In some instances, states only apply the 0.1% HFR concentration limit to the sale of certain goods, including children's products, residential upholstered furniture, textiles, mattresses, and/or bedding. Maine currently has the most extensive restrictions on BFRs, banning the sale of all commercial goods containing any quantity of any BFR, as well as the sale of residential upholstered furniture containing HFRs above 0.1%. It is notable that for those states which have not specified “all commercial goods”, the range of goods listed can differ significantly. For example, Minnesota specifies that HFR content should be restricted (to 0.1%) in children's products, upholstered furniture, and mattresses. Rhode Island's limit of 100 mg/kg for all HFRs meanwhile does not include children's products but specifies that bedding and furniture are to be restricted. Finally, Maine limits HFR content to 1,000 mg/kg in residential upholstered furniture yet has no specific mention of bedding/mattresses, and New Hampshire (also 1,000 mg/kg in upholstered furniture) explicitly excludes mattresses from inclusion in its regulation. While individual states have enacted limits on hazardous flame retardants, the lack of a federal mandate may result in the purview of these laws differing significantly (California Legislature et al., 2003, Hawaii State Legislature, 2004, Maine Legislature, 2004, New York Legislature and Marcellino, 2004, Illinois Legislature et al., 2005, Maryland Legislature, 2005, Michigan Legislature et al., 2005, Oregon Legislature et al., 2005, Minnesota Legislature, 2007, Washington Legislature, 2007, Oregon Legislature et al., 2009, Vermont Legislature and House Committee on Health Care, 2009, Maryland Legislature, 2010, Vermont Legislature et al., 2013, Minnesota Legislature et al., 2015, Washington Legislature, 2016, Maine Legislature, 2017, Rhode Island Legislature, 2017, California Legislature et al., 2018, Minnesota Legislature et al., 2019).

4.3. Asia-Pacific Region

The major e-waste and textile producers in the Asia-Pacific Region

Table 2

EU Regulations and Directives governing POP-BFRs mixtures along with accompanying concentrations limits for articles that are entering waste streams or entering the market, i.e. concentrations above which an article cannot enter the designated stream. (a) Does **not** apply to electrical and electronic equipment (EEE) within the scope of Directive 2011/65/EU (RoHS) (b) Applies **solely** to EEE within the scope of the RoHS directive. (c) Does **NOT** include the deca-BDE congener. (d) “Other Wastes” include that from thermal processes, C&D (bricks, mortar, soil, etc.), waste water treatment plants, and waste management processes (see text of Regulation 2019/1021 for more details). (e) This listing includes the sum of all congeners with the same degree of bromination but separate from other congeners, e.g. all penta-BDE congeners, but none of tetra-BDE congeners, etc. * See Supplementary Information – S3 for definitions of Substance, Mixture, and Article.

EU Regulation/Directive	BFR(s)	Concentration Limit Imposed by EU Regulation/Directive		
		Waste	Market (UTC)	Other Wastes ^(d)
2019/1021 (POPs)	HBB	0.005%	0	0.5%
2019/1021 (POPs)	HBCDD	0.1%	0.01%	0.1%
2019/1021 (POPs)	tetra-, penta-, hexa-, hepta-, or deca-BDE ^(e)	<i>not included</i>	0.001% ^(a) (substance*)	<i>not included</i>
2019/1021 (POPs)	Σ(tetra-, penta-, hexa-, hepta-, and deca-BDE)	0.1%	0.05% ^(a) (mixture/article*)	1% ^(c)
2011/65/EU (RoHS)	SPBBs	<i>not included</i>	0.1% ^(b)	<i>not included</i>
2011/65/EU (RoHS)	SPBDEs	<i>not included</i>	0.1% ^(b)	<i>not included</i>
1907/2006 (REACH)	Deca-BDE	<i>not included</i>	0.1%	<i>not included</i>

have adopted all of the Stockholm Convention annexes which relate to POP-BFRs (Table 3), with the exception of India who have only ratified the Stockholm Convention with regards to the initial 12 POPs (Sharma, 2014, UNEP, 2020e). However, India have reportedly banned the trade, use, import and export of HBB, HBCDD, commercial Penta-BDE, and commercial Octa-BDE, and have also implemented 1,000 mg/kg concentration limits on certain electrical and electronic equipment, following some of the provisions of the EU’s RoHS Directive (Chetry, 2018). Indonesia meanwhile has ratified the annexes to the treaty, though no national policies or regulations have been established with regards to concentration limits in consumer or waste goods. Both China and Japan have meanwhile carried out extensive research and implemented strategies for the curtailing of these chemicals, in line with Stockholm Convention provisions.

In 2016, the Standing Committee of the National People’s Congress of China “... banned the produce, use, import, and export of HBCDD except for specific exemptions of production and use for EPS and XPS in buildings”, in line with exemptions set out in the Stockholm Convention (Jiang et al., 2017). China has also established concentration limits for PBDEs (the congeners related to the Penta- and Octa-BDE commercial mixtures) of 0.1% in “electronic information products” made from virgin plastics entering the marketplace (Wang and Luo, 2006, Ren et al., 2017). However, they have yet to include a limit on Deca-BDE in said materials likely due to its only recent listing in the Stockholm Convention and the economic burden of implementing screening protocols and finding replacement flame retardants. There is likewise no specific legislation regulating the unintentional presence of FRs in recycled

Table 3

Dates of entry into force of amendments to annexes A, B, or C to the Stockholm Convention for major e-waste or textile producing states. (a) Currently availing of an exemption pursuant to the use of the relevant chemical/mixture in certain circumstances. (b) Currently availing of an exemption pursuant to the production of the relevant chemical/mixture in certain circumstances.

	HBB	Hexa/Hepta-BDEs	Tetra/Penta-BDEs	HBCDD	Deca-BDE
EU	26-08-10	26-08-10	26-08-10 ^(a)	22-04-16	18-12-18
China	26-13-14	26-03-14	26-03-14	26-12-16 ^(a,b)	none
USA	none	none	none	none	none
Japan	26-08-14	26-08-14	26-08-14	26-11-14	18-12-18
India	none	none	none	none	none
Brazil	26-08-10	26-08-10 ^(a)	26-08-10 ^(a)	26-11-14 ^(a)	18-12-18 ^(a)
Indonesia	26-08-10	26-08-10	26-08-10	26-11-14	18-12-18

plastic materials (Cao et al., 2019).

Japan has no history of production or use of HBB and also ceased all use of commercial Octa-BDE in the 1990s (UNEP, 2017b). Though there are currently no concentration limits on the presence of HBB or other BFRs in the majority of consumer articles, they have classified HBB, along with tetra-, penta-, hexa-, and hepta-BDEs, as Class I Specified Chemical Substances which virtually prohibits the manufacture, import, and use of these chemicals in all applications (Ministry of the Environment, 2016). A notable exception to this are

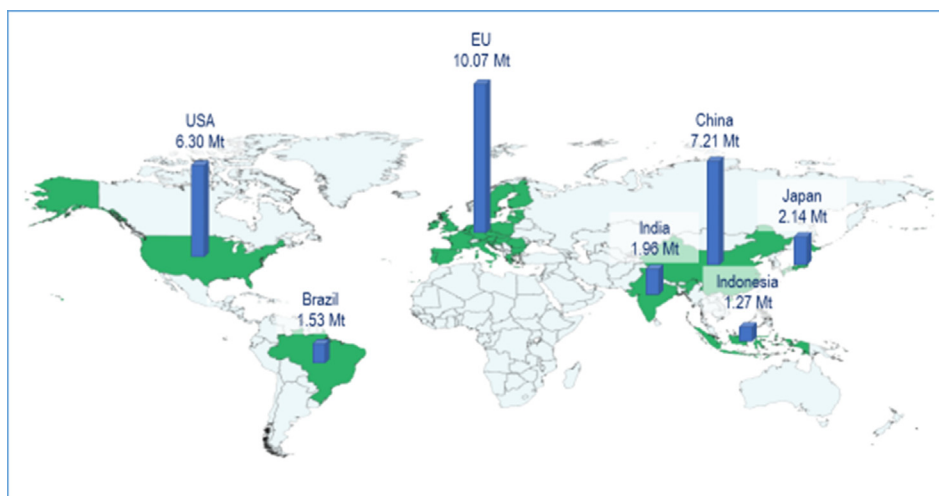


Fig. 2. Largest e-waste producers worldwide in 2016 (values provided given in megatons (Mt)/10⁶ tonnes – adapted from Baldé et al, 2017).

Table 4

List of US states which have Bills in place that regulate the concentrations of brominated/halogenated flame retardants in various commercial goods, with a brief overview of said Bills' purview and concentration limits.

US State	Chemicals	Restricted Goods	Concentration Limit (mg/kg)	Date
California	Penta-/Octa-BDE	All Commercial Goods	1,000	01/06/06
	HFR	Child's products, mattresses, upholstered furniture	1,000	01/01/20
Hawaii	Penta-/Octa-BDE	All Commercial Goods	1,000	01/01/06
Illinois	Penta-/Octa-BDE	All Commercial Goods	1,000	01/01/06
Maine	All BFRs	All Commercial Goods	None	01/01/10
	HFRs	Residential Upholstered Furniture	1,000	01/01/19
Maryland	Penta-/Octa-BDE	All Commercial Goods	1,000	01/10/08
	Deca-BDE	Mattresses, Residential Upholstered Furniture, EEE	None	31/12/10
Michigan	Penta-BDE	All Commercial Goods	1,000	01/06/06
Minnesota	Penta-/Octa-BDE	All Commercial Goods	1,000	01/06/06
	Deca-BDE, HBCDD	Children's Products, Residential Upholstered Furniture	1,000	01/07/19
	HFRs	Children's Products, Upholstered Furniture, Business/Residential Textiles, Mattresses	1,000	01/07/19
New York	Penta-/Octa-BDE	All Commercial Goods	1,000	01/01/06
New Hampshire	HFRs	Upholstered Furniture	1,000	08/05/19
Oregon	Penta-/Octa-/Deca-BDE	All Commercial Goods	1,000	01/01/06
Rhode Island	Penta-/Octa-BDE	All Commercial Goods	1,000	01/01/07
	HFR	Upholstered Bedding & Furniture	100	01/07/19
Vermont	Penta-/Octa-BDE	All Commercial Goods	1,000	01/07/09
	Deca-BDE	Mattresses, Mattress Pads, Upholstered Furniture, Plastic Housing from TVs/Computers	1,000	01/07/19
Washington	Penta-/Octa-BDE	Non-Edible Commercial Goods	None	01/01/08
	Deca-BDE	Mattresses, Residential Upholstered Furniture, Plastic Housing from TVs/Computers	1,000	01/01/11
	Deca-BDE, HBCDD	Children's Products, Residential Upholstered Furniture	1,000	01/07/17

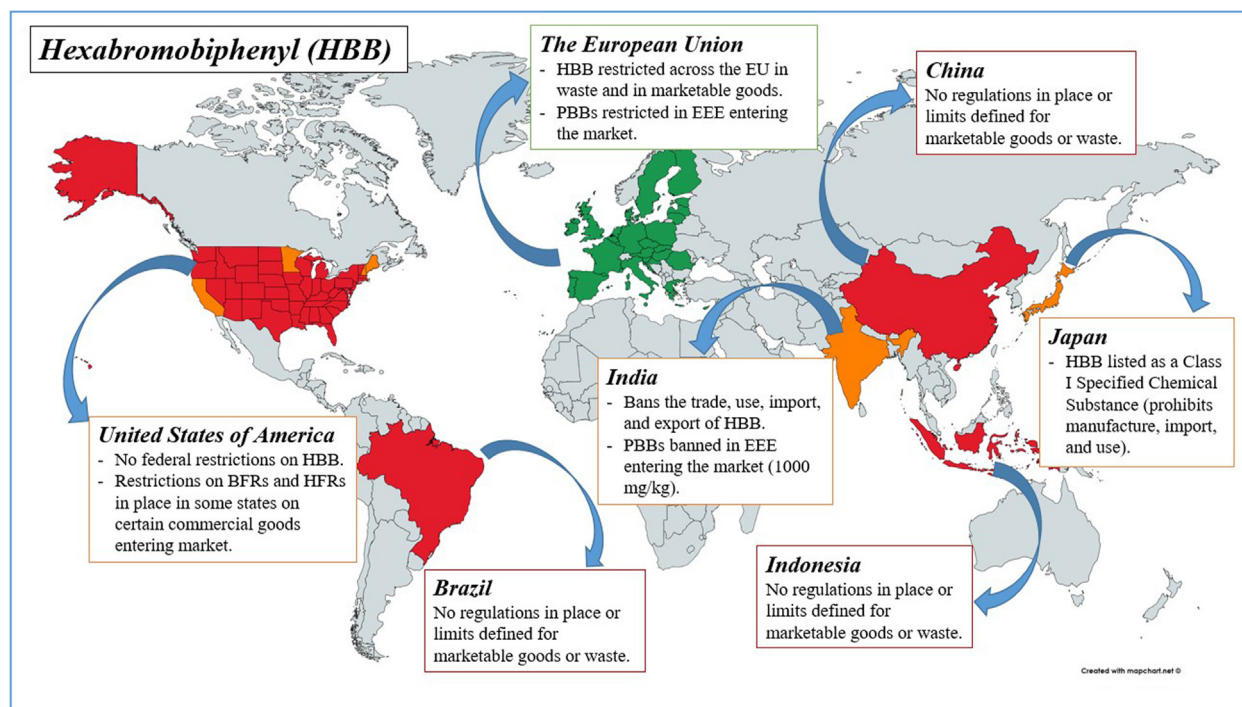


Fig. 3. Colour-coordinated maps reflecting the extent to which the major waste-producing countries (Fig. 2) have adopted measures to prohibit the use and re-use of POP-BFRs: Green (restrictions on relevant POP-BFR in goods entering the market, entering the waste stream, and with defined concentrations limits); Orange (some official measures in place to reduce relevant POP-BFR presence and/or use); Red (no restrictions currently in place for the relevant POP-BFRs). See Section 3 for in-depth information on EU policies, and Section 4 for information on policies in other countries worldwide. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

limits on the sum of PBBs and PBDEs (at 1,000 mg/kg) in certain electrical and electronic equipment. As outlined in Sections 2.2 and 2.3, Japan is currently registered for the exemption to recycle cathode ray tubes (CRTs) containing PBDEs and is closely monitoring the application of HBCDD in insulation materials and seeking alternatives for the eventual prohibition of the chemical (Ministry of the Environment, 2016) (overview of worldwide BFR restrictions shown in Fig. 3).

5. Discussion

5.1. Phasing-out of legacy BFRs

With the recent restrictions on the use of Deca-BDE, the Stockholm Convention now encapsulates many of the major legacy BFR mixtures used in EEE, vehicles, textiles, and building materials over the last few decades. Through agreement to this MEA, states and REIOs that are party to the Convention are required to closely monitor and restrict the

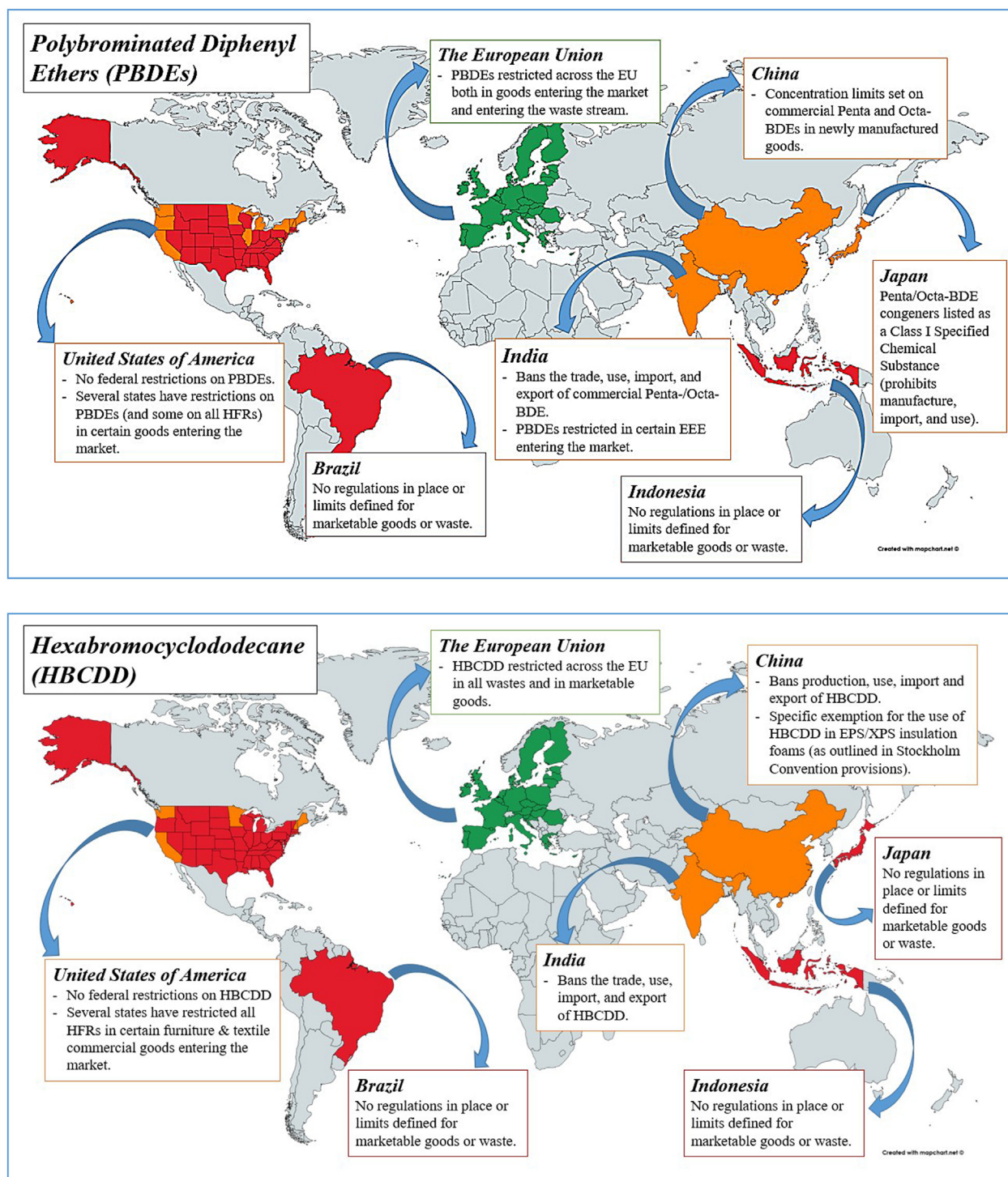


Fig. 3. (continued)

use of these chemicals within their jurisdictions. The relevant major waste producing countries outlined in this review have each pushed forward on these restrictions to varying degrees within their own economic areas. This has been most extensively undertaken by the EU by overseeing three key sectors where articles containing hazardous BFRs could be detected, controlled, and/or removed from circulation: in articles manufactured (at least in part) with potentially contaminated materials; in newly-manufactured articles made from virgin plastics entering the consumer market; and in end-of-life articles entering the waste stream (EU, 2019). The ideal scenario is the removal of all traces of these hazardous chemicals from articles both currently in and

entering into circulation. However, such a situation is not practically feasible due to the economic impact such an endeavour would have, as well as the workload involved in screening the huge volume of consumer articles either manufactured or entering the waste stream (Dragé et al., 2018).

Allowances such as these are foreseen and taken into consideration by the Stockholm Convention for certain applications of the listed chemicals. The Convention is, in a large sense, voluntary, and the list of specific exemptions gives a degree of breathing space to parties, thus allowing a more smooth transition to a commercially viable alternative to the restricted chemical. Deference is also given to parties with

developing economies as more pressing issues such as poverty and famine will inevitably be prioritised over environmental conservation; a sentiment which is reflected by the first principle of the Rio Declaration, one of the cornerstones upon which the Stockholm Convention is established (UN, 1992). Such allowances let countries such as China and Japan – who have ratified most, if not all, of the provisions of the Convention – to create restrictions as is within their current capabilities, restrictions which will likely develop in line with those the EU have passed.

It is noteworthy that certain states within the US have gone even further than what has been outlined by UNEP and the Stockholm Convention. States such as California, Maine, Minnesota, New Hampshire, and Rhode Island have restricted all halogenated flame retardants in selected commercial goods, and a few have even gone so far as to restrict the use of all chemical flame retardants in certain commercial goods. However, in contrast to EU policy (which aligns most EU states' regulations) and those of other large countries such as China and India, the disparity between the states' regulations may create other issues. The extent of transboundary movement of goods across state lines means that many manufacturers and retailers may be forced to default to the most stringent laws to ensure they meet legal requirements. However, the lack of congruent legislation nationwide also means that goods manufactured and sold within state lines may still contain these hazardous substances. There are also no restrictions on the management of waste containing BFRs in the US, nor is there in any of the major waste producing countries outlined in [Section 4.1](#) (with the exception of the EU). Furthermore, the technology to screen and remove said chemicals is not yet implementable at industrial scales (Hennebert and Filella, 2018; Harrad et al., 2019). This then creates an issue for the recycling of contaminated plastics, the recirculation of hazardous BFRs in new consumer articles, and their subsequent export across state lines and even worldwide.

The practical and cost-effective methods for preventing further environmental contamination from these POPs for developed states and REIOs are perhaps exemplified by those restrictions currently placed by the EU on its member states. Those restrictions and the accompanying exemptions must to be tailored to each individual POP to account for factors such as relative toxicity, ubiquity, and suitability of replacement chemicals, as well as the economic capability of the state. For instance, the use of HBB as a flame retardant was commonplace in the United States until a well-documented agricultural mishap involving contamination of livestock feed with the chemical (Eggington, 1980). This led to much investigation into the biological and environmental hazards posed by this and other similar chemicals both within the US and worldwide, though many manufacturers and utilisers of HBB began to voluntarily phase out its use in the following years (Eljarrat and Barceló, 2011). Thus the large-scale use of HBB ceased by the late 1970s and the low concentrations found now in consumer and waste goods is reflected in the restrictions put in place by the EU: a limit of 50 mg/kg HBB in articles entering the waste stream – in consideration of older articles contaminated by proximity to HBB-treated articles – and no allowed UTC limit for newly manufactured goods (EU, 2019).

Penta-BDE and Octa-BDE, meanwhile, were in circulation and in use until much more recently, a point which is similarly reflected by the conditions of the Stockholm Convention and EU regulations. With articles which may have been treated with these mixtures potentially still in circulation, the Convention makes an allowance specifically for the recycling of goods containing those chemicals, provided reasonable oversight and management of the newly-made product is carried out. Therefore, the imposition by the EU of a strict UTC of 500 mg/kg for the sum of PBDEs in articles entering the market gives an allowance for recyclable waste containing unknown quantities of Penta- and Octa-BDE-treated articles while still imposing a limitation on the volumes re-entering circulation. Similar allowances are made for HBCDD, with a UTC of 100 mg/kg for newly-manufactured articles and a limit of 1,000 mg/kg for waste (EU, 2019). An allowance for the use of HBCDD

in building materials was also made while a viable alternative was sought; for example, the sole use of polymeric flame retardants (poly-FRs) in extruded and expanded polystyrene insulation foams in Germany (Schlummer et al., 2015; Koch et al., 2016).

Deca-BDE has many more specific exemptions compared to the less brominated PBDEs and HBCDD, as expected due to it being the most recent addition to the Convention's list of POPs. As Deca-BDE was allowed and used in many applications until very recently, reasonable time and allowances are available to those states and REIOs which require its continued use without enduring undue economic burdens. The EU's previous limit of up to 1,000 mg/kg deca-BDE in articles placed on the market gave leeway to goods made from recycled materials. The recent recast of the EU POPs Directive has begun to lower that limit, with a single UTC for all PBDEs in articles entering the market. This limit will subsequently be revised and likely lowered come the July 2021 revision date (EU, 2019).

5.2. Limitations of concentration limits

Though the adoption of concentration limits for hazardous BFRs in certain consumer articles has led to the decline in the use of said BFRs worldwide, this has been accompanied by an increase in the use of NBFRs as replacements. This can be seen as a limitation to the effectiveness of the Stockholm Convention and subsequent legislations. Alternatives to outlawed chemicals will enter the market in order to meet flammability standards for certain goods, while those replacements may themselves be deemed hazardous in the future owing to the similar chemical properties to their banned predecessors (Charbonnet et al., 2020). The recent listing of Deca-BDE in the Convention and its subsequent ban in the EU has led to the rise in use of decabromodiphenyl ethane (DBDPE) (UNEP, 2008b; Wemken et al., 2020; Harrad et al., 2020), an NBFR which is structurally very similar to Deca-BDE (Eljarrat and Barceló, 2011). Similarly, the recent ban on the use of HBCDD in building insulation foams has led to a replacement with so-called "Poly-FRs", high molecular weight butadiene styrene BFRs now currently in use by major insulation manufacturers worldwide (Global Insulation, 2014; Global Insulation, 2016). Poly-FRs are suggested to be a safer alternative to HBCDD, as they are chemically bound to the polymers to which they are added making it much more difficult to leach out. These new additives are the foci of much research currently due to their increasing prevalence and their chemical properties which are similar to those that led their predecessors to be listed as POPs (Blum et al., 2019). However, no legislation currently exists specific to these replacement compounds, which can hinder screening methods being developed for the detection and removal of POP-BFRs (Harrad et al., 2019). This is where some of the more recent US state directives can be said to have superiority, as some have proactively included all halogenated flame retardants, or even all flame retardants, in their restrictions. These are likely responses to the revising of the California Technical Bulletin on flammability in upholstered furniture which, among other effects, requires much less or no chemical flame retardants to meet reasonable fire safety standards (State of California, 2013). Though these state laws themselves are limited in many cases to certain commercial goods (mainly upholstered furniture), their broad mandates preclude the use of alternative replacement flame retardants where none may actually be required.

The management of BFR-treated waste is another noteworthy area which requires careful consideration. Concentration limits are carefully selected to consider unintentional contamination of BFRs in newly-manufactured goods and downstream contamination in goods made from recycled materials. These limits must also factor in concentrations of BFRs typically found in goods to be recycled and the environmental hazards they pose at said concentrations. This complex set of criteria result in concentration limits typically around 0.001–0.1%. Though these limits are in place, the technology to actively screen waste or consumer articles is not suitable in many cases due to the accuracy

required to effectively screen the waste as well as the sheer volume of goods which may potentially contain hazardous substances. So-called “gold-standard” techniques such as GC-MS and LC-MS would prove unfeasibly slow and expensive to operate at this scale, though the accuracy and precision of the analysis could be virtually guaranteed. There are, however, more rapid alternatives being developed, such as portable X-Ray Fluorescence (XRF) or Fourier-Transform Infra-Red (FTIR) technologies. Despite neither being able to identify the species of BFR present in a sample and both requiring sizable margins of error, specific methodologies have been detailed which demonstrate their screening potential to a high degree of certainty (Schlummer et al., 2015, Hennebert and Filella, 2018, Sharkey et al., 2018, Wagner and Schlummer, 2020). However, both have other considerations for the required large-scale use. FTIR is an “on-line” system, not necessarily requiring many operators for its daily use, but the instrument itself is much more expensive compared to portable XRF instrumentation and can also only accurately quantify to roughly 1% bromine content, well above even the highest restrictions of BFR-content in consumer articles (Turner and Filella, 2017, Sharkey et al., 2018). XRF methods would likely require many operators to be trained in the use of the instrument to physically screen the waste and also requires a measurement time of 30–60 s per sample, resulting in a large work-force and potentially several instruments per site to screen effectively and efficiently. However, the XRF has a comparatively much lower limit of detection; as low as 0.001%, well within the levels required for screening for current concentration limits (Gallen et al., 2014). The use of XRF coupled with implementation of current POP-BFR concentration limits was reported to be over 90% effective in screening waste plastics in a large-scale study (Harrad et al., 2019). Such a screening method allows for (as much as practically feasible) the removal of BFR-treated articles from circulation while still allowing the majority of “clean” materials to be recycled – the vast majority of screened waste articles not containing BFRs above concentration limits. Improvements are also constantly being made to these technologies and methodologies which may further improve their screening potential and viability. However, the use of non-regulated BFRs can then undermine the effectiveness of these instruments, limiting their ability to screen for compliance within defined concentration limits. Besides which, these technologies or similar screening methods are not currently in place in many jurisdictions which have defined BFR concentration limits. Therefore, current concentration limits in jurisdictions without effective screening methods in place mainly prevent the further use of hazardous BFRs in newly manufactured goods but likely do little to prevent the re-circulation of BFRs in recycled materials.

Questions must ultimately be raised about the necessity for using flame retardants so ubiquitously in consumer articles in the first instance. The expansion of current legislation to encompass more (or even all) flame retardants is a hotly debated topic (de Boer and Stapleton, 2019b). Advocates for the continued use of flame retarding chemicals maintain that the use of such additives is the most effective way to meet flammability standards (BSEF, 2018, ChemicalSafetyFacts, 2020). Meanwhile, other groups suggest that generalising the vast array of flame retarding chemicals as a single class to be prohibited is misguided, as they may have differing chemical properties and some may be relatively safe for use (Osimitz et al., 2019). However, the revision of the California Technical Bulletin in 2013 has shown that fire safety regulations can be updated to better reflect modern consumer habits (State of California, 2013). Current data strongly suggests that factors such as decreases in smoking rates, increased use of smoke detectors and improved fire-safety education played a much larger role in the improvement of fire-safety standards over recent decades (Ahrens, 2019, Charbonnet et al., 2020). Additionally, there has been a clear pattern of flame retardants being regulated, only to be replaced by another similar compound, which in turn was found to be of concern, in a repeating cycle of regrettable substitutions (Eljarrat and Barceló, 2011, de Boer and Stapleton, 2019a). Much time and resources are

devoted to identifying and regulating these hazardous substances, by which time replacements are already likely in circulation. More priority should then be given to revising flame retardancy standards such that the requirement for potentially harmful and persistent substances is curtailed in the future. This would ideally break the seemingly unending cycle of hazardous flame retardant use and act a guideline to other families of persistent organic chemicals which see similar cycles occurring.

6. Conclusions

Treaties such as the Stockholm Convention and the resulting legislation created by its signatories are merely the first step in removing hazardous chemicals such as POPs from circulation. From this, research and regulations must work together to find practical ways to remove POPs from circulation and use the knowledge garnered to forewarn for similar environmental issues which may occur in the future. Much research is being undertaken to establish ways to detect and remove legacy brominated flame retardants and other such known hazards from consumer goods. However, the use of NBFRs and alternative flame retardants will likely result in an unending cycle of new and hard to remove chemical additives being introduced, removed, and replaced by newer additives that may in turn prove to be hazardous. While work to remove persistent chemicals currently in use is vital (particularly for chemicals so resistant to natural degradation), focus is also needed on the requirement to use these chemicals in the first place. The US states who have begun to restrict the use of *all* flame retardants in certain applications have taken a major step forward in one regard, revising the need for flame retardancy in common commercial goods (following California TB 117-2013). However, the lack of congruent legislation countrywide and worldwide, as well as the technology to screen and remove said chemicals not yet implementable at industrial scales, renders such bold steps only partially effective. The Stockholm Convention attempts to harmonise the most prevalent hazardous chemicals which can be dealt with unanimously worldwide, in an attempt to clean up what is currently adversely affecting the environment. Alongside the Convention, technological innovations to effectively screen for those pollutants must be developed and coupled with congruent legislative actions worldwide in order to reduce the need for using such chemicals in the future.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2020.106041>.

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